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Soft x-ray magnetic circular dichroism study of Mn–Ir/Co–Fe bilayers with giant exchange anisotropy

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Soft x-ray magnetic circular dichroism (XMCD) and element-specific magnetic hysteresis (ESMH) measurements were performed in transmission mode on $Mn_{73}Ir_{27}$ 10 nm/$Co_{70}Fe_{30}$ 2 nm bilayers with different chemical orderings of the Mn–Ir layer.The unidirectional anisotropy constant was 0.55 erg/cm^2 for the disordered Mn–Ir layer and 1.18 erg/cm^2 for the ordered Mn–Ir layer. The XMCD signal of Mn was observed, which means the induced ferromagnetic component of Mn spins through the exchange coupling at the interface. No vertical offset of the Mn ESMH loops was observed for either the disordered or the ordered bilayers, which means that insignificant uncompensated Mn spin was pinned at the interface to induce exchange bias on the Co–Fe layer.
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Exchange anisotropy[1,2] is an indispensable physical phenomenon for the realization of high-density magnetic storage devices such as hard disk drives (HDDs) and magnetic random access memories (MRAMs). Recently, giant exchange anisotropy, exceeding 1 erg/cm^2 in the unidirectional anisotropy constant J_K, has been observed in Mn–Ir/Co–Fe bilayers containing an $L1_2$-Mn_3Ir phase.[3] This has made it possible to further reduce the dimensions of the spin valve elements in HDDs and MRAMs. However, the microscopic origin of the giant exchange anisotropy has not yet been clarified. In order to understand the mechanism of exchange anisotropy in terms of the spin structure, neutron scattering[4,5] and x-ray magnetic circular/linear dichroism[5–9] are currently used for ferromagnetic (FM)/antiferromagnetic (AFM) bilayer systems. X-ray magnetic circular dichroism (XMCD) is a powerful tool for detecting uncompensated AFM spins, making it possible to determine the magnetization process of AFM spins accompanied by a reversing FM moment. Ohldag *et al.* reported on the correlation between exchange bias and pinned interfacial uncompensated AFM spins through XMCD studies on several exchange biased bilayer systems.[10] They concluded that the observed exchange anisotropy energy (J_K) is dominated by the ideal interfacial coupling energy J between the FM and the AFM layers, according to a simple extension of the Meiklejohn and Bean model.[11,12] The experimental fact they deduced the above conclusion from is that the pinned interfacial uncompensated AFM spin is always 4% of the interface layer, regardless of the material combinations of the bilayer system, which can show different exchange bias strengths. Their thesis is thus examined when the vertical offset of a hysteresis loop of uncompensated AFM spins, which corresponds to the pinned interfacial uncompensated spins, is investigated for bilayer systems having the same J and different J_K values. From this viewpoint, the role of pinned uncompensated spins in yielding an exchange bias will be well characterized in a $Mn_{75}Ir_{25}$/Co–Fe system because the J_K is enhanced remarkably when the chemical ordering of Mn and Ir atoms is promoted.[3] Since the atomic ordering of a Mn–Ir layer does not change the face centered cubic crystallographic structure and the chemical composition at the interface,[13] it is implied that the ideal interfacial coupling energy J is constant against the enhancement of J_K with ordering. In the present study, therefore, we used XMCD to measure element-specific magnetic hysteresis (ESMH) loops of Mn–Ir/Co–Fe bilayers with different chemical orderings of the Mn–Ir layers to clarify the correlation between the vertical offset of the hysteresis loop and the exchange biasing strength.

The specimens were deposited on 100-nm-thick Si-N membrane substrates with a Si-N/$Cr_{66}Ni_{24}Fe_{10}$(5)/Ru(20)/$Mn_{73}Ir_{27}$(10)/$Co_{70}Fe_{30}$(2)/Ru(1)/$Cr_{66}Ni_{24}Fe_{10}$(1) design (thickness units in nanometers) by magnetron sputtering, according to the methods in Refs. 3 and 14. For the purpose of structural characterization, identically structured films were also fabricated on thermally oxidized Si wafers. The substrates were held at room temperature (RT) during the deposition, except for that of the Mn–Ir layer. In order to change the chemical ordering of the Mn–Ir layer, two different substrate temperatures T_{sub} were employed (RT and 170 °C).[3] After breaking the vacuum, the specimen fabricated at T_{sub}=RT (or 170 °C) was annealed under a vacuum pressure less than 5×10^{-6} Torr, at the optimal condition of 280 °C (360 °C) for 1 (5) h and was then cooled to RT, in order to induce the largest J_K for the respective specimens.[14]

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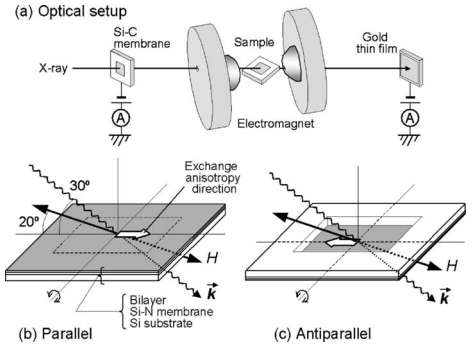

FIG. 1. (a) Experimental geometry used for the acquisition of XMCD and ESMH. In order to achieve (b) near parallel and (c) near antiparallel configurations between the exchange anisotropy direction and the x-ray wave vector $\boldsymbol{k}$, the sample was turned over so x rays penetrated from either (b) the film side or (c) the membrane side.

During the annealing procedure, an external magnetic field of 1 kOe was continuously applied.

The microstructures of the specimens were examined by x-ray diffraction and grazing incidence x-ray diffraction (GID). The degree of order S of the Mn–Ir layer was determined using the integral intensity ratio of the Mn_3Ir (110) and (220) peaks observed in the GID profiles.[15] Since the Mn–Ir (220) diffraction peak was superimposed on the Ru (110) diffraction peak, these peaks were separated by referring to the GID profile for a Cr–Ni–Fe/Ru bilayer.[14] The intensity ratios, $I_{(110)}/I_{(220)}$, were 0 and 0.24 for T_{sub}=RT and 170 °C, respectively, while it is 0.79 for perfectly ordered Mn_3Ir with (111) preferred orientation. Since S is proportional to the square root of $I_{(110)}/I_{(220)}$, S was determined to be 0 and 0.55 for T_{sub}=RT and 170 °C, respectively. Hereafter we refer simply to the bilayer with S=0 (T_{sub}=RT) as the disordered bilayer and that with S=0.55 (T_{sub}=170 °C) as the ordered bilayer.

X-ray absorption spectra (XAS) were obtained by recording the x-ray intensity ratio of the transmission to the incidence as a function of x-ray energy, which differs from the method used by Ohldag *et al.*, where the total electron yield (TEY) of specimens was measured.[10] Compared to the TEY method, the transmission method or reflection method[5] is less sensitive to artifacts owing to the magnetic field applied to specimens, and so is favorable for studying weak MCD signals from uncompensated AFM spins. MCD was obtained using a 1 Hz helicity switching technique with left and right circularly polarized x rays with a degree of circular polarization of $p=\pm 0.96$ from the twin helical undulators at BL25SU of SPring-8.[16] Figure 1 shows a schematic illustration of the experimental geometry. The ESMH of Co (Mn) was obtained by recording the difference in MCD between the L_3 and L_2 edges as a function of the applied field strength, where x-ray energies were 779.1 eV (639.9 eV) and 794.5 eV (652.5 eV), respectively.

Figure 2 shows the XAS at the Mn $L_{2,3}$ and Co $L_{2,3}$ edges measured for both the disordered and the ordered bilayers. A small offset of the base lines of the MCD spectra is an artifact from the geometrical setup of the spectrometer. Small spatial deviation of x-ray beams at the sample surface and also at the intensity monitoring membrane/film, owing to the different points of generation of photons with the respective helicities, might explain this artifact.

For both bilayers, we can clearly see the MCD signals not only at the Co $L_{2,3}$ edges but also at the Mn $L_{2,3}$ edges.

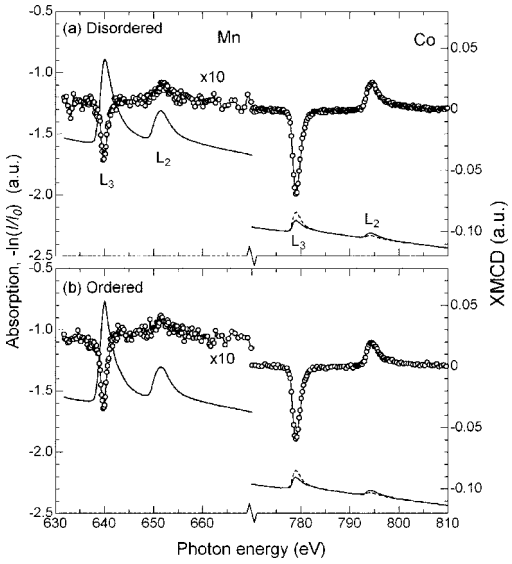

FIG. 2. XMCD and absorption spectra at the Mn $L_{2,3}$ and Co $L_{2,3}$ edges in the (a) disordered and (b) ordered Mn–Ir/Co–Fe bilayers acquired at room temperature. Open circles joined by a solid line correspond to XMCD spectra. Solid and broken lines represent absorption spectra for the plus and minus helicities, respectively, of the incident photons. A magnetic field of +14 kOe was applied during the acquisition.

On the other hand, when we measured XAS for a Mn–Ir film without an adjacent Co–Fe layer [CrNiFe(5)/Ru(20)/Mn–Ir(5)/Ru(1)/CrNiFe(1)], the MCD at the Mn $L_{2,3}$ edges was not observed at all. This fact means that uncompensated Mn spins, in other words, ferromagnetic components of Mn magnetization, are induced in the bilayer through the exchange coupling at the interface between the FM and the AFM layers and are not due to defects in Mn films such as surface steps, lattice vacancies, and grain boundaries. Namely, it is a purely interfacial effect. In fact, the uncompensated AFM moments steeply decrease within 3 ML apart from the interface, according to the Heisenberg model calculation.[17] The same sign for the Mn MCD signal and the Co MCD signal indicates the parallel configuration between the FM spins and the uncompensated Mn spins. By applying the magneto-optical sum rule[18] to the absorption and MCD spectra, we can estimate the magnitude of uncompensated Mn spins. To avoid an extrinsic influence from the offset of the MCD base lines, the difference spectrum between the MCD spectra measured at +14 and −14 kOe was employed for analysis. For instance, in the case of a disordered bilayer, the uncompensated Mn moment is about $0.02\mu_B$ per atom and less than 1% of the atomic moment of Mn ($2.5\mu_B$) in bulk $Mn_{75}Ir_{25}$ alloys.[19] When we assume that the uncompensated Mn spins are localized at the interfacial monolayer of the AFM layer, the estimated magnitude corresponds to 40% of the total atomic moment of Mn at the interface and is comparable to the value ($52\pm13\%$) reported in Ref. 10.

Figure 3 shows the Mn and Co ESMH loops for (a) the disordered and (b) the ordered bilayers. In order to eliminate the offset of MCD base lines in the spectra mentioned above, the ESMH loops are calculated using the MCD at both the L_3 and the L_2 edges, according to Ref. 10. Since the sample magnetization contributes the opposite sign and the artificial offset contributes the same sign to the asymmetry at the L_3 and L_2 edges, we simply took the difference between them ($MCD_{L3}-MCD_{L2}$) and plotted this as a function of the ex-

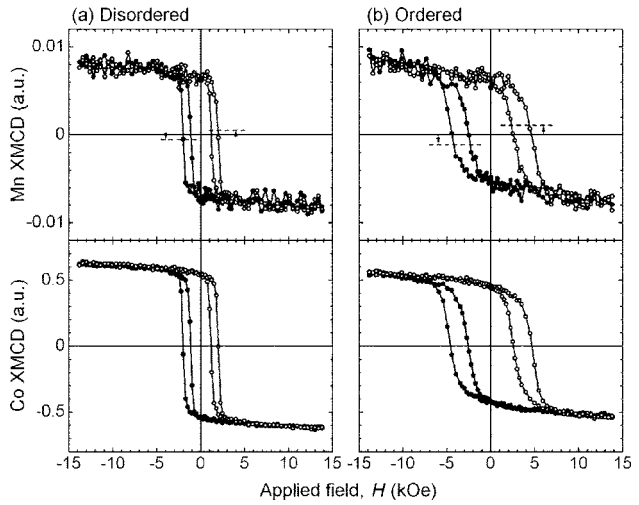

FIG. 3. Mn and Co ESMH loops of the (a) disordered and (b) ordered Mn–Ir/Co–Fe bilayers recorded at room temperature. The hysteresis loops were acquired with the exchange bias direction either parallel (solid circles) or antiparallel (open circles) to the incident x-ray wave vector. Vertical offsets of Mn ESMH loops are not observed. Dashed lines with small arrows indicate the expected vertical offsets, assuming that (a) 7% and (b) 14% of the total uncompensated Mn moments are pinned.

ternal applied field strength. This procedure is indispensable for distinguishing between the contribution of pinned spins to the MCD in ESMH and the artificial offset of the MCD signal, because neither responds to the magnetic field. The vertical axes of the figure are normalized with the averaged x-ray absorption intensity at the respective L_3 peak. The saturation MCD amplitude of Mn ESMH is almost the same between the disordered and the ordered bilayers.

We will now discuss the exchange bias phenomena. In both bilayers, the Co ESMH loops exhibit a typical horizontal loop shift. The Mn ESMH loops also exhibit a horizontal shift, and their shapes and horizontal shifting fields correspond well with those of the respective Co ESMH loops. The averaged shifting field is 1.58 kOe for the disordered bilayers and 3.60 kOe for the ordered bilayers. Using the separately determined $M_s d_F$ value of the Co–Fe layer in the simultaneously deposited specimens on the Si wafer, these shifting fields are converted into J_K ($\equiv M_s d_F H_{ex}$) values of 0.55 and 1.18 ergs/cm^2, respectively. When we follow the thesis of Ohldag *et al.* expressed by the equation $J_K=\rho J$,[10] assuming a constant J, the doubling of J_K in the ordered bilayer in comparison with that in the disordered one must originate from a twofold pinned fraction ρ of uncompensated interfacial AFM spins. This means that the vertical offset of the Mn ESMH loop of the ordered bilayer must be twice that of the disordered one. However, as shown in Fig. 3(a), one cannot see any vertical offsets in either the parallel or the antiparallel Mn loops, within the accuracy of the present experiment. If there were a pinned fraction of about 7% of the total uncompensated Mn moments as Ohldag *et al.* observed,[10] the Mn hysteresis loops should be separated from each other by 0.0011 in the vertical axis scale of Fig. 3. While the signal to noise ratio is not great, a separation of 0.0011 along the vertical axis would be detected. Furthermore, as can be seen in Fig. 3(b) for the ordered bilayer, no vertical offset of the Mn ESMH is observed, similarly to the disordered case. This experimental fact means that all the uncompensated Mn moments follow the rotation of Co moments, and insignificant amounts of pinned fractions of uncompensated Mn moments are present in both the disordered and the ordered bilayers. Of course, any magnetic asymmetry against the field reversal is needed to raise the exchange anisotropy. Therefore, according to the present experimental results, we should take into account the asymmetry of the compensated AFM spin structure that is not detected by the MCD technique to elucidate the mechanism of exchange anisotropy. One possible asymmetry of the compensated AFM spin structure is the formation of the interfacial domain wall in the AFM layer. While the uncompensated AFM moments at the interface follow the FM spin reversal as we observed, the compensated AFM spins far inside the AFM layer will not follow this. Because of the exchange stiffness between AFM spins, the twisted compensated AFM spin structure will be formed in the AFM layer.

In summary, soft x-ray MCD was measured in transmission mode for $Mn_{73}Ir_{27}$ 10 nm/$Co_{70}Fe_{30}$ 2 nm bilayers with different chemical orderings of the Mn–Ir layer. No vertical offset of the Mn ESMH loops was observed for either the disordered or the ordered bilayer. We conclude that the thesis of Ohldag *et al.*, in which the pinned interfacial Mn spins dominates the exchange bias strength, is not applicable to the giant exchange anisotropy of Mn–Ir/Co–Fe bilayers with a chemically ordered Mn–Ir layer.

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